

APPENDIX B:

Modeled Outdoor Concentrations of Hazardous Air Pollutants:

Analysis of Data from the Cumulative Exposure Project

for the Urban Area Source Program

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1 INTRODUCTION

In order to gain an expanded understanding of the national distribution of air toxics concentrations, EPA's Office of Policy, Planning and Evaluation has conducted a national air toxics modeling study as part of its Cumulative Exposure Project. Outdoor concentrations of 148 air toxics were modeled at the census tract level for the entire continental U.S., in both urban and rural areas. To evaluate the potential impacts of air toxics in urban areas, modeled concentrations were compared with potential regulatory thresholds of concern or "benchmark concentrations." Modeled concentrations greater than these benchmark concentrations provide an indication of a potential health risk to the general population. The frequency and magnitude of modeled concentrations greater than benchmark concentrations provide an indication of those hazardous air pollutants having the greatest potential to pose health risks to the general population.

This chapter presents the methods for and results of this screening-level study of national urban air toxics concentrations. In addition, results for urban areas are compared with those for rural areas. Contributions of area sources of emissions, relative to point sources and mobile sources, are also assessed.

2 METHODS

There are three primary components to this analysis of urban air toxics:

1. Estimating Air Toxics Emissions and Modeling Air Toxics Concentrations
2. Identifying Benchmark Concentrations
3. Comparing Modeled Concentrations to Benchmark Concentrations.

The methods used in each of these components are discussed below.

2.1 Estimating Air Toxics Emissions and Modeling Air Toxics Concentrations

2.1.1 Atmospheric Modeling Methodology

To meet this study's objectives of estimating long-term concentrations of HAPs on a national scale with reasonable geographic resolution, a long-term Gaussian dispersion modeling approach was selected. The Assessment System for Population Exposure Nationwide (ASPEN) used in this study is a modified version of EPA's Human Exposure Model (HEM). The HEM is a standard modeling tool used by EPA staff to support regulatory activity and special studies, such as EPA's electric utility study (U.S. Environmental Protection Agency 1996d). The HEM utilizes a Gaussian dispersion modeling approach for point sources with optional first-order decay and a simple deposition algorithm. For this study, the deposition algorithm has been improved and treatment of area and mobile source emissions has been modified. In addition, methods to estimate secondary formation of seven HAPs (formaldehyde, acetaldehyde, propionaldehyde, methyl ethyl ketone, acrolein, phosgene, and cresol) have been incorporated.

For each source, the model calculates ground-level concentrations as a function of radial distance and direction from the source for a set of receptors laid out in a radial grid pattern. These concentrations represent the steady-state concentrations that would occur with constant emissions and meteorological parameters, and are calculated for model receptor locations up to 50 kilometers (km) from each source. Concentrations resulting

from any number of sources are extrapolated from model receptor locations to the centroids of population subdivisions, such as census tracts, block groups, or blocks. The model may be used to simulate any size modeling domain for which appropriate data are available.

In recognition of the potential for a large degree of spatial variation in ambient concentrations, geographic resolution at the census tract level was selected for this study. There are approximately 60,000 census tracts in the continental United States. Census tracts generally contain roughly equal populations, and therefore tend to be smaller in urban areas and larger in rural areas. This level of resolution focuses greater computing resources in urban locations, and appropriately balances the desire for high spatial resolution against the limitations of the model and the available emissions and meteorological databases that preclude accurate modeling at higher resolution. The modeled concentrations approximate the population-weighted average of outdoor HAP concentrations experienced within a census tract over the course of a year.

Modeling was conducted separately for each source category group identified in the following section. For each HAP, the modeled concentrations for each category group, along with the background concentrations identified below, were summed together to arrive at a modeled concentration for each census tract. Details of the ASPEN model are found in Chapter 5 of the technical report on the modeling portion of this study (Rosenbaum et al. 1998).

2.1.2 Emissions Inventory

2.1.2.1 Emissions categories and data sources

HAPs are emitted from a variety of sources, including large manufacturing facilities such as chemical production plants, combustion facilities such as waste incinerators, small commercial operations such as dry cleaners, and both onroad and nonroad mobile sources. For this analysis, sources of HAP emissions were aggregated into six source category groups:

- manufacturing point sources (e.g., chemical manufacturing, refineries, primary metals)
- nonmanufacturing point sources (e.g., electric utility generators, municipal waste combustors)
- manufacturing area sources (e.g., wood products manufacturing, degreasing)
- nonmanufacturing area sources (e.g., dry cleaning, consumer products, small medical waste incinerators)
- onroad mobile sources (e.g., cars, buses, trucks)
- nonroad mobile sources (e.g., farm equipment, airplanes, boats).

In this study, emissions from manufacturing point sources are represented by data from EPA's 1990 Toxics Release Inventory (TRI) (U.S. Environmental Protection Agency 1991). TRI is an annual compilation of facility-reported estimates of releases and transfers for over 300 pollutants. TRI is a comprehensive database of estimated emissions for large manufacturing sources, but does not address many important sources of toxics emissions, including mobile sources, combustion sources such as incinerators, and small industrial, commercial, consumer and agricultural sources (i.e. area sources). For these other source

category groups, this study estimates HAP emissions from EPA's extensive 1990 base year national emissions inventories for volatile organic compounds (VOCs) and particulate matter (PM) (Pechan 1994; U.S. Environmental Protection Agency 1993b). These inventories contain facility-specific data on point sources and county-level emissions totals for mobile sources and area sources.

HAP emissions estimates are derived from VOC and PM emission estimates through the application of speciation profiles. Speciation profiles are industry-specific and/or process-specific estimates of the presence of particular chemical constituents in a VOC or PM emissions stream. For example, estimates of gaseous HAP emissions from automobile refinishing operations can be derived by combining the estimated total VOC emissions from automobile refinishing with a speciation profile, which provides estimates of the percentage of automobile refinishing VOC emissions made up of individual chemical constituents, such as benzene and toluene. Speciation profiles are available from EPA's SPECIATE database (U.S. Environmental Protection Agency 1992) and from the technical literature (Battye and Williams 1994; Burnet et al. 1990; California Air Resources Board 1991; Edgerton et al. 1985; Hare and White 1991; Harley and Cass 1994; Harley et al. 1992; Hildemann et al. 1991; Ingalls 1991; Lipari et al. 1984; Miller et al. 1994; Sagebiel et al. 1996; Scheff et al. 1992; Scheff et al. 1994; U.S. Environmental Protection Agency 1989; U.S. Environmental Protection Agency 1996d). Speciation profiles are not available for all source categories; for categories without profiles, emissions are estimated using profiles for source categories judged to be similar. This introduces some uncertainty into the characterization of emissions. However, it is

necessary to use estimates with uncertainties in order to approximate actual HAP concentrations, since many sources and source categories have not been characterized in detail. To omit such sources could result in significant underestimates of HAP concentrations.

In this analysis, the definition of an area source is not precisely equivalent to the CAA Section 112 definition, nor is the definition of a point source precisely equivalent to the CAA section 112 definition of a major source. Area source emissions in this analysis are derived from EPA's national inventories of VOC and PM emissions. In these inventories, area sources are defined as those that do not emit more than 100 tons per year of any criteria pollutant (VOCs, PM, carbon monoxide, nitrogen oxides, or sulfur dioxide). Some of the emissions in this analysis attributed to area sources may actually be emitted by facilities which are defined as major sources under CAA section 112—that is, facilities which emit more than 10 tons per year of any single HAP or 25 tons per year of any combination of HAPs. There also may be facilities classified as point sources in this analysis which are considered area sources under CAA section 112. The potential implications of these different definitions of area sources are discussed in Section 4 below.

The scope of the modeling exercise was limited to a certain extent by the available emissions data. The domain for the modeling exercise is comprised of the continental United States. The national VOC and PM inventories used to develop much of the HAP emissions estimates for this study do not include data for Alaska or Hawaii; these states were therefore not included in the modeling study. In addition, 148 out of the 188 HAPs

listed in CAA section 112(b)(1) are included in the modeling study. There were no emissions data identified for the remaining HAPs in the data sources used.¹

2.1.2.2 Spatial Allocation

For point sources, emissions data are available for specific facility locations. All emissions data for area sources and mobile sources, however, are estimated as county totals. Before HAP emissions derived from these inventories can be used effectively in dispersion modeling, they should be allocated to smaller geographic units within the counties to approximate the spatial distribution of actual emissions.

To allocate emissions from the county level to the census tract level, 20 different surrogates were developed, as shown in Table 1. Each surrogate is based on data available at the census tract level, and represents an approximation of the distribution of emissions-generating activities across the census tracts within a county for at least one area source or mobile source category of emissions. Surrogates were developed using data on population (Bureau of the Census 1990a; Bureau of the Census 1990b), roadway miles and railway miles (Bureau of the Census 1993), and land use (U.S. Geological Survey various dates) for each census tract. For each area source and mobile source category, county emissions were allocated to constituent census tracts in proportion to the fraction of the total county value of the corresponding surrogate. For example, each county's emissions from lawn and garden equipment are allocated to each census tract in

¹ A few HAPs with available emissions data, such as chlorine and titanium tetrachloride, were not included in the modeling exercise because of their physical/chemical properties which make dispersion modeling difficult.

proportion to the tract's percentage of residential land area within the county, while emissions from industrial sources are allocated in proportion to industrial land use, and mobile source emissions are allocated using data on population and roadway miles in each tract.

2.1.3 Background Concentrations

The emissions inventory and modeling methodologies described above are used to estimate long-term outdoor concentrations of HAPs attributable to 1990 anthropogenic emissions, within 50 kilometers of each source. For many HAPs, however, outdoor concentrations may include "background" components attributable to long-range transport, re-suspension of historical emissions, and non-anthropogenic sources. To accurately estimate 1990 outdoor concentrations of HAPs, it is necessary to account for these background concentrations which are not represented by atmospheric modeling of 1990 anthropogenic emissions.

In this study, background concentrations are represented by inclusion of concentration values measured at "clean air locations" remote from the impact of local anthropogenic sources. Background concentrations were identified from the literature (Grosjean 1991; Howard 1989; Howard 1990; Howard 1991; Howard 1992; Howard 1997; Khalil and Rasmussen 1984; Panshin and Hites 1994a; Panshin and Hites 1994b; Singh et al. 1985; U.S. Environmental Protection Agency 1994a; U.S. Environmental Protection Agency 1996c; Wiedmann et al. 1994; World Meteorological Organization 1991) for 28 HAPs, and are shown in Table 2. These values were obtained from measurement studies

characterized as rural/remote, hemispheric baseline, remote ocean, global background, or other terms denoting contributions from only natural sources or long-range transport. For these HAPs, the estimated concentration in each census tract is determined by the summing together the estimated background concentrations and the modeled concentrations arising from 1990 emissions. Background concentrations are assumed to be constant across all census tracts; the available data are insufficient to address any possible geographic variations in background. Because no background concentration values were identified in the technical literature for other 120 HAPs included in this study, their background concentrations are assumed to be zero. This may result in underestimation of outdoor concentrations for some HAPs.

2.1.4 Evaluation of Modeled Concentrations

Performance of the model was evaluated by comparing modeled concentrations of HAPs to available monitored concentrations. In addition, modeled and monitored carbon monoxide (CO) concentrations were also compared for evaluation of model performance. Although CO is not a HAP, it was included in the model simulations specifically for model evaluation purposes, because the CO measurement data base contains significantly more monitoring sites than the HAP measurement data base. In terms of dispersion and other atmospheric processes after release, CO is expected to behave similarly to gaseous HAPs with very low reactive decay rates. In addition, CO is measured hourly throughout the year, whereas HAP measurements are typically 24-hour averages taken approximately every twelfth day. The greater temporal coverage reduces uncertainty in annual average statistics, and allows for time-of-day comparisons.

2.1.4.1 Monitoring Data Sets

Quantitative comparisons of 1990 annual average ASPEN model predictions with observed HAP concentrations were made for several monitoring programs summarized in Table 3. Some of the programs were not operating in 1990, so measurements for other years between 1988 and 1992 were used in some cases, introducing some uncertainty into the comparisons.

HAP measurement data from the monitoring programs identified in the table were not used for the quantitative model performance evaluation in any cases where more than 10 percent of the measurements were below the minimum detection levels (MDLs). The uncertainty in estimating an annual average concentration from monitoring data sets with large numbers of non-detects limits the usefulness of such data sets in quantitative model performance evaluation. More qualitative analysis of some of these data sets was used to supplement the information from the quantitative model performance evaluation. For data sets used in the quantitative analysis, those values below the MDL were set equal to half the detection limit as a default in calculating annual average concentrations.

CO data from 259 monitoring sites were extracted from EPA's Aerometric Information Retrieval System (AIRS) for comparison with ASPEN predictions. In selecting the sites, an attempt was made to eliminate those monitors identified as microscale or middle-scale and/or as maximum concentration or source-oriented. Because these monitors are located

in order to detect extreme concentrations, or “hot spots”, they are likely to record concentrations that are significantly higher than the ASPEN estimates for the corresponding census tracts, which represent tract averages. However, not all monitor records contained these identifiers, and some are likely to be incorrectly identified. Therefore, a certain amount of underprediction of CO concentrations for this set of 259 monitor locations is expected. A separate analysis was conducted for the 100 CO monitors out of the set of 259 that were specifically designated as representative of neighborhood scale (0.5 to 4 km), urban scale (4 to 50 km), or regional scale (more than 50 km). Comparison of model outputs with measured concentrations from this subset of monitors provides an analysis that is not influenced by any unidentified “hot spot” monitors included in the larger set of 259.

2.1.4.2 Evaluation Methods

For each HAP at each monitoring location with a full year of data and fewer than 10 percent of observations below the MDL, the ratio of the predicted (modeled) concentration to the observed (monitored) concentration was calculated. In order to account for the possibility that a pollutant monitor may be nearly equidistant from multiple census tract centroids, measured concentrations were compared with a distance-weighted average of the nearest six tract concentration predictions, weighting each centroid value by $1/\text{distance}^2$. Note that the ASPEN algorithms are designed to estimate concentrations that represent the average throughout the census tract. Although the monitored HAP values are point measurements, they are typically made in locations where concentration gradients are not expected to be steep, because the long-term monitoring programs from

which they are taken are intended to represent general population exposures. Sensitivity analysis showed that results are not substantially different when only the closest census tract is used for comparison.

The same evaluation methods were also used for the CO model-monitor comparisons. An assumed CO background concentration of 125 ppb (144 $\mu\text{g}/\text{m}^3$) was added to the sum of predicted anthropogenic contributions to CO concentrations for this comparison. This value is based on 1989-1990 measurements at Niwot Ridge, Colorado (Novelli et al. 1992), a remote land site at approximately intermediate U.S. latitude (40N).

2.2 Benchmark Concentrations

Toxicological information on health effects and dose-response relationships for the 148 hazardous air pollutants included in this study was assembled from a variety of sources, evaluated comparatively, and assigned to a series of tiers defined by quality and availability of information, and consistency of methodology. Much of the needed health effects information was previously compiled for EPA's proposed rulemaking under the authority of section 112(g) of the Clean Air Act. For this study, the 112(g) information was updated, and information from several additional data sources was incorporated (Caldwell et al. 1998).

Hazard information and dose-response data for the HAPs were used to develop benchmark concentrations for carcinogenic hazard and for noncarcinogenic hazard from both long-term and short-term exposure. For each hazard category, a benchmark

concentration representing a presumptive health protective value was selected. For carcinogenic hazard, the benchmark was selected to be the concentration of a known, probable, or possible human carcinogen representing an upper bound one-in-one-million excess probability of contracting cancer over a lifetime of exposure. The selection of this benchmark is based on provisions of CAA sections 112(c)(9) and 112(f) that allow source categories to be exempted from regulation when posing less than a one-in-one-million lifetime cancer risk to the most exposed individual.

For noncarcinogenic hazards, the benchmark was selected to be the concentration of a HAP likely to be without appreciable risk of noncancer effects from long-term or short-term exposures. This benchmark is based on the provision of section 112(f) of the CAA requiring “residual risk” emissions standards to “provide an ample margin of safety to protect public health” from non-cancer effects. Similar language is also found in CAA section 112(c)(9). EPA inhalation reference concentrations (RfCs), or similar values developed by other agencies, representing levels below which long-term exposure is not expected to result in any adverse health effects, were selected as the benchmark concentrations for non-cancer health effects from long-term exposure.

Benchmark concentrations for potential non-cancer hazards from short-term exposures to HAPs were developed using EPA’s Levels of Concern (LOC). LOCs are established for chemicals on the Superfund Amendments and Reauthorization Act section 302 list of “extremely hazardous substances” (U.S. Environmental Protection Agency et al. 1987). LOCs are airborne concentrations of chemicals for which no serious irreversible health

effects are expected to occur following a short-term exposure of 30 minutes. To derive a short-term benchmark concentration, the LOC was divided by a safety factor of 1000 to address the fact that the LOC is based on lethality as an endpoint and to address the uncertainty in the derivation of the LOC.

Wherever available, EPA estimates of inhalation unit risks (IURs) for cancer and EPA reference concentrations (RfCs) for non-cancer effects were used in developing benchmark concentrations. When these values were not available, other available values were used as benchmark concentrations, including: EPA estimates of cancer risks from oral exposure, converted into inhalation units; California EPA inhalation unit risks and Reference Exposure Levels (RELs); and Minimal Risk Levels (MRLs) developed by the Agency for Toxic Substances and Disease Registry (ATSDR).

Fourteen of the 148 HAPs included in this study are chemical groups. It is difficult to assess the toxicity of chemical groups because each is comprised of a number of different constituents that may have varying levels of toxicity. For this analysis, toxicity values that can be assigned to an entire chemical group are included (Caldwell et al. 1998). Toxicity values applicable only to individual constituents of chemical groups are not included, because the modeled concentrations developed in this study represent the entire group.

The various benchmark concentrations were grouped into tiers to account for differences in methodology, inherent uncertainty of data used in derivation, and level of peer review. Tier I includes values derived from EPA IURs and RfCs, and represents those values with

the most consistency in derivation and highest level of peer review. Tier II includes other categories of EPA data, as well as quantitative information from California EPA and ATSDR.

2.3 Comparison of Modeled Concentrations to Benchmark Concentrations

This study's modeled concentrations are estimates of annual average outdoor HAP concentrations for 1990. To screen for whether a modeled concentration represents a potential health risk, it is compared to benchmark concentrations for cancer and chronic noncancer effects, assuming long-term exposure. These benchmark concentrations represent an estimated concentration at which a lifetime daily exposure is unlikely to result in adverse health effects, based on available hazard assessment data. A modeled long-term concentration greater than a cancer or chronic benchmark is therefore an indicator of some potential for adverse health effects.

In addition, estimated ambient concentrations are also compared to benchmarks for health effect concerns from short-term exposure. While the estimated concentrations in this analysis do not represent short-term peak concentrations typically used to assess acute effects, exceedance of short-term benchmarks by long-term average concentrations is an indication of a potential health concern, because short-term peak concentrations will be higher than annual average concentrations. However, the uncertainties in the benchmarks for short-term exposure are relatively large.

Comparison of estimated HAP concentrations to benchmark concentrations implicitly treats outdoor concentrations as equivalent to exposure concentrations. Outdoor concentrations are a reasonable proxy for exposures that occur both outdoors and indoors, given the high rates of penetration into indoor environments for various HAPs. A field sampling study of indoor and outdoor concentrations of volatile organic compounds (VOCs) reported by Lewis (Lewis 1991) and Lewis and Zweidinger (Lewis and Zweidinger 1992) found that penetration of VOCs from outdoor to indoor air is complete, even when air exchange rates are low. Similar results have been found for particulates less than 10 micrograms in aerodynamic diameter--that is, the penetration of such pollutants from outdoor to indoor air is virtually 100 percent (U.S. Environmental Protection Agency 1996a). Therefore, long term indoor concentrations of both gaseous and particulate HAPs can, in the absence of indoor sources, be assumed to be approximately equal to long term outdoor concentrations in the same location. Indoor removal mechanisms may reduce indoor concentrations to some extent for some HAPs.

To evaluate the potential for individual HAPs to pose health risks, hazard ratios were computed by dividing each estimated HAP concentration by its corresponding benchmark concentration for both cancer and noncancer health effects. Hazard ratios greater than one indicate the estimated concentration was in excess of the benchmark concentration. Hazard ratios were computed for each available benchmark for each census tract.

To evaluate the impacts of air toxics in urban areas, exceedances of benchmark concentrations are evaluated separately for urban census tracts and rural census tracts.

Each census tract was designated as either “urban” or “rural” as part of the dispersion modeling methodology, since dispersion parameters differ for these two types of locations. All census tracts with population density greater than 750 people/km² are designated as urban, while census tracts with lower population density are designated as rural, based on EPA modeling guidance (U.S. Environmental Protection Agency 1996b). This results in an approximately even split of census tracts into the urban and rural designations, meaning that many suburban areas are classified as “rural” rather than “urban.” Characteristics of urban and rural census tracts are shown in Table 4.

3 RESULTS

3.1 HAP Modeling

3.1.1 Pollutant Concentrations

Figure 1 shows boxplots of the modeled concentrations of 38 selected HAPs in the 28,314 urban census tracts. The HAPs shown are those identified in section 3.3 below as having at least 50 urban census tracts with modeled concentrations exceeding health hazard benchmark concentrations. Pollutants are separated into four groups in the figure, according to the magnitude of the modeled concentrations shown on the vertical axes. The vertical lines for each HAP in the figure show the range from the 1st percentile to the 99th percentile of modeled concentrations, while the box shows the range from the 25th to the 75th percentile, and the horizontal line shows the median modeled concentration for urban census tracts. Modeled concentrations of zero were set equal to a minimal value (1×10^{-13}) to accommodate a logarithmic scale. In the fourth panel of the figure, pollutants for which the modeled concentration is zero in a majority of the urban census tracts are shown with a median concentration equal to this minimal value.

For many of the HAPs shown, the distribution of modeled concentrations spans several orders of magnitude; however, the range from the 25th to the 75th percentile is one order of magnitude or less for a majority of the pollutants. Greater variations in the modeled concentrations are seen for pollutants that have a relatively large proportion of emissions from point sources, such as propylene dichloride and hydrazine.

3.1.2 Model Performance

Table 5 summarizes predicted-to-observed concentration ratios for all HAPs for which a significant amount of monitoring data above the minimum detect level was identified. All available observed data from the monitoring programs listed in Table 3 were combined for each HAP. The results for these HAPs show an overall tendency for the model to underestimate the observed concentrations, with geometric mean predicted/observed ratios generally less than 1.0, ranging from 0.09 to 1.0.

P-dichlorobenzene, methylene chloride, and styrene appear to be underpredicted to a greater degree than other gaseous HAPs, with geometric mean predicted-to-observed ratios less than 0.33, suggesting that significant sources have been omitted from the emissions inventory for these pollutants.

Model-monitor comparisons for CO also indicate a tendency for underestimation of concentrations. For the full set of 259 monitors, the geometric mean ratio is 0.52, and for the subset of 100 monitors specifically identified as not related to “hot spots,” the geometric mean ratio is 0.62. Further analysis of the predicted-to-observed ratios for CO, described in Chapter 7 and Attachment 5 of the modeling report (Rosenbaum et al. 1998) suggests that much of the model’s tendency to underestimate pollutant concentrations may be explained by limitations of the Gaussian model formulation, such as neglect of calm wind conditions, poor representation of stable atmospheric conditions, and the 50 kilometer downwind distance limit. Uncertainties in the HAP emissions inventory may also explain the tendency to underpredict.

Figure 2 presents a comparison of predicted and observed concentrations for 13 gaseous HAPs at 5 locations in the city of Baltimore, Maryland. The overall Spearman correlation coefficient is 0.82 ($p=0.00001$). The high correlation coefficient is an indicator of good performance in distinguishing the relative magnitude of concentrations among the different HAPs included in the data set.

3.2 Benchmark Concentrations

A total of 183 benchmark concentrations were identified for the 148 HAPs in this study, as summarized in Table 6. Seventy-seven of the benchmarks are for cancer, 90 for non-cancer effects from long-term exposure, and 16 for non-cancer effects from short-term exposure. No quantitative benchmarks were identified for 29 of the 148 HAPs, while benchmarks for more than one of the three hazard categories were identified for 60 HAPs.

Out of the 14 pollutant groups included in this study, benchmarks appropriate for applications to the entire group were identified for nine: arsenic compounds, beryllium compounds, cadmium compounds, chromium compounds, cobalt compounds, lead compounds, manganese compounds, nickel compounds, and selenium compounds. For five pollutant groups, no benchmarks applicable to the entire group were identified for antimony compounds, cyanide compounds, glycol ethers, mercury compounds, and polycyclic organic matter. Values of all benchmark concentrations used in this analysis are shown in Attachment 1.

3.3 Benchmark Exceedances

3.3.1 Exceedances of Benchmark Concentrations in Urban and Rural Census

Tracts

Table 7 shows the percentage of urban and rural census tracts with modeled 1990 average outdoor concentrations that exceed benchmark concentrations. Results are shown for 38 HAPs with exceedances in more than 50 urban census tracts². An additional 12 HAPs not shown in the table have exceedances in between 10 and 50 urban census tracts, and an additional 17 have exceedances in between 1 and 10 urban census tracts. Three pollutants have exceedances in rural census tracts only; in each case, the number of census tracts is less than five.

A majority of the benchmark concentrations exceeded are for cancer, rather than noncancer effects. This reflects the fact that the cancer benchmarks, set at a one-in-one-million risk level, are generally much lower concentrations than the noncancer benchmarks.

There are eight HAPs (benzene, bis (2-ethylhexyl) phthalate, carbon tetrachloride, chloroform, ethylene dibromide, ethylene dichloride, formaldehyde, and methyl chloride) with benchmark exceedances in all urban and rural census tracts. For each of these HAPs, the background concentration alone, as defined in section 2.1.3 above, exceeds the benchmark concentration for cancer, as shown in Table 8. To evaluate the impact of 1990

² Modeled concentrations for many of the HAPs may exceed more than one benchmark, as 60 HAPs have more than one benchmark concentration (separate benchmarks for cancer, non-cancer effects from long-

anthropogenic emissions, the background concentration was subtracted from the total estimated concentrations, and the remaining concentrations (i.e., modeled 1990 anthropogenic concentrations) were compared to benchmark concentrations, with results also shown in Table 8. Modeled 1990 anthropogenic concentrations for benzene and formaldehyde are estimated to exceed the benchmark concentration in almost all urban census tracts and in more than 87 percent of rural census tracts. Modeled anthropogenic ethylene dichloride concentrations exceed the benchmark concentration in 32 percent of urban census tracts and 11 percent of rural census tracts. The remaining five HAPs have 1990 modeled anthropogenic concentrations that exceed benchmark concentrations in less than 10 percent of both urban and rural census tracts.

For most other HAPs in Table 7, the percentage of urban census tracts with exceedances of benchmark concentrations is two to three times greater than the percentage for rural census tracts for most pollutants. For some pollutants, however, including 1,3-dichloropropene, p-dichlorobenzene and lead, the relative frequency of exceedances in urban areas is much greater. For example, modeled concentrations of 1,3-dichloropropene exceed the cancer Tier II benchmark for this pollutant in 56 percent of urban census tracts and in only 5 percent of rural census tracts. Almost all estimated emissions of this pollutant are associated with consumer lawn care products. National emissions estimates from this category are spatially allocated to census tracts in proportion to residential population, resulting in greater emissions density in urban tracts. For this category, this allocation method may overstate the share of emissions in urban census

term exposure, and non-cancer effects from short-term exposure). In Tables 7-12, HAP results are shown

tracts relative to rural census tracts. P-dichlorobenzene and lead also have large proportions of emissions that are allocated to tracts in proportion to population.

Of the pollutants with more than 50 urban exceedances, only benzotrachloride has a higher frequency of exceedances in rural tracts than in urban tracts. Virtually all of the estimated benzotrachloride emissions used in this analysis were reported to the TRI by four facilities. Two of these facilities, accounting for about 68 percent of the reported emissions, are in rural locations.

3.3.2 Relative Contributions of Area, Point and Mobile Sources

To evaluate the contribution of three broad categories of sources—area sources, point sources, and mobile sources—to exceedances of benchmark concentrations in urban areas, the estimated number of exceedances resulting from each category's emissions was calculated separately. That is, the modeled concentrations associated with area source emissions were compared to the benchmark concentrations for each HAP in each census tract, and the same was done for point source and mobile source emissions.

Area source emissions estimates were developed for 73 of the HAPs included in this analysis. Table 9 lists the HAPs for which area source emissions alone are estimated to result in long-term outdoor concentrations greater than the benchmark concentrations. There are 30 HAPs with modeled exceedances of benchmark concentrations resulting from estimated area source emissions. Six HAPs—acrolein, benzene, 1,3-butadiene, 1,3-

only for the benchmark with the most exceedances for each HAP.

dichloropropene, formaldehyde, and chromium—are estimated to have concentrations greater than the benchmarks, from area source emissions alone, in more than 90 percent of urban census tracts. Four of the HAPs shown in the table are estimated to have area source exceedances in less than 0.1 percent of urban census tracts.

Point source emissions have been estimated for all 148 HAPs included in this analysis. There are 63 HAPs with benchmark exceedances in urban census tracts resulting from estimated point source emissions alone; 34 of these have exceedances in more than 50 urban census tracts and are shown in Table 10. In general, point source emissions of individual HAPs tend to result in exceedances in a smaller number of census tracts than area sources: only seven HAPs—arsenic, benzene, 1,3-butadiene, dioxin, chromium, formaldehyde, and nickel—are estimated to exceed the benchmark concentration in ten percent or more of the urban census tracts due to point source emissions alone. Each of these HAPs, except dioxin, also has a large number of exceedances from area source emissions alone.

Mobile source emissions were estimated for 35 HAPs in this analysis. As shown in Table 11, ten HAPs exceed benchmark concentrations in urban census tracts when only mobile source emissions are considered. Five of these HAPs—acrolein, benzene, 1,3-butadiene, chromium, and formaldehyde—have modeled mobile source concentrations that exceed benchmarks in more than 90 percent of urban census tracts, while acetaldehyde is estimated to exceed its cancer benchmark in two-thirds of the urban tracts from mobile source emissions alone. The results shown in Table 11 for particulate HAPs are

attributable to estimated emissions from diesel vehicles (for arsenic, chromium, and nickel, based on speciation data in the technical literature (Hildemann et al. 1991; and commercial marine vehicles powered by residual oil (for cadmium and nickel).

The results presented in Tables 9, 10 and 11 consider the impacts of area, point and mobile sources in isolation. For some HAPs, only one of these source category groups appears to account for most of the benchmark exceedances, while for other HAPs the contributions of two or all three of the source category groups are very significant. Table 12 compares the number of estimated exceedances from modeled concentrations for each of the three broad source category groups alone (from Tables 9-11), along with exceedances for all source categories and background concentrations combined (from Table 7). This comparison is shown for each of the 38 HAPs with estimated exceedances in more than 50 urban census tracts. HAPs are listed in order of the number of benchmark exceedances resulting from modeled area source emissions only.

Table 12 shows that modeled concentrations of 1,3-butadiene from area sources alone exceed the cancer benchmark concentration for this HAP in almost every urban census tract, and that the same is also true of modeled mobile source concentrations for this HAP. This suggests that emissions from both area sources and mobile sources would have to be reduced in order to realize urban concentrations of 1,3-butadiene that are lower than this HAP's cancer benchmark concentration. This finding also applies to acrolein, benzene, formaldehyde, and chromium compounds, since each of these HAPs also exceed benchmark concentrations from modeled area sources alone and from modeled mobile

sources alone in nearly every urban census tract. Each of these pollutants also has smaller, but still significant, contributions from point sources. A different finding applies to 1,3-dichloropropene. Modeled concentrations of this HAP from area sources alone (consumer lawn care products) also exceed the benchmark concentration in nearly every urban census tract, but there are no estimated emissions of this HAP from mobile sources, and minimal contributions from point sources.

Table 12 also shows that vinyl chloride and p-dichlorobenzene each have modeled concentrations from estimated area source emissions alone that exceed benchmark concentrations in more than 35 percent of urban census tracts. Both pollutants have smaller estimated contributions from other source types. For both of these pollutants, however, the number of benchmark exceedances resulting from area source emissions may be overstated due to uncertainties in the emissions inventory; estimated area source chemical manufacturing emissions of these pollutants have high uncertainties and may be overestimates.

The benchmark exceedances for most of the other pollutants in Table 12 appear to be due to the combined contributions of area sources and point sources, with significant contributions for some HAPs from mobile sources (nickel compounds, arsenic compounds, and particularly acetaldehyde) and from background concentrations. For the last nine HAPs listed in the table, benchmark concentrations are exceeded in fewer than 500 hundred urban census tracts each, and these exceedances are due almost entirely to emissions from point sources.

A correlation analysis of the number of benchmark exceedances for each source category group and the total number of exceedances, for the set of HAPs in Table 12, shows that exceedances from area sources are most strongly correlated with total exceedances. The relationship between the exceedances from area sources and the total, while controlling for mobile sources and point sources, is a correlation of 0.68 ($p=0.0001$). The correlation between the exceedances from the point sources and the total, while controlling for the area and mobile sources is 0.48 ($p=0.0001$). The relationship between the exceedances from mobile sources and the total, while controlling for the area and point sources, is a correlation of 0.14 ($p=0.0001$). This indicates that the exceedances from area sources have the strongest association with total exceedances, and that there is also an important contribution from point sources. Mobile source exceedances do not appear to be associated with total exceedances when considering the full set of HAPs in Table 12. As discussed above, mobile source contributions are very important for several individual HAPs; but for HAPs with large mobile source contributions, there are also large contributions from area sources.

4 LIMITATIONS AND UNCERTAINTIES

The analysis presented in this chapter uses available emissions data, modeling techniques and hazard evaluation data to estimate the frequency with which long-term average concentrations of hazardous air pollutants at the census tract level may be greater than benchmark concentrations. While modeled concentrations are subject to uncertainties arising from both emissions estimates and the modeling methodology, the available

monitoring data support the conclusion that exceedances of benchmark concentrations are common. For example, several sources of long-term monitoring data for benzene and 1,3-butadiene show that measured concentrations routinely exceed benchmark concentrations (California Air Resources Board 1995; New York State 1993; Texas Natural Resource Conservation Commission 1997).

4.1 Emissions Estimates

The majority of HAP emissions estimates used in this study were developed through the application of speciation profiles to the 1990 base year national interim emissions inventories for VOCs (1993 version) and particulate matter (PM) (1995 version). The speciation methodology starts with a large data set—national emissions of total VOCs and PM—and breaks it down into relevant component parts, i.e. emissions of each of the HAPs. The strength of this approach is in its comprehensive coverage of sources and in applying a consistent methodology nationally. Uncertainties in this approach are due to uncertainties in the VOC and PM inventories and in the speciation profiles used to estimate the HAP components of the VOC and PM emissions.

Another approach to estimating national HAP emissions for a modeling study is to assemble a variety of different data sources that each address a particular set of HAP emitters (source categories) or a particular geographic area. The strength of this approach is that it frequently has more detailed emissions estimation methodologies. Uncertainties in this approach are due to possible inconsistencies that are introduced by combining data from disparate sources, and the potential for overlooking important emitters.

An important goal for the modeling portion of this study was to approximate actual outdoor concentrations of air toxics in 1990. Therefore, key objectives included comprehensive treatment of emissions and emissions sources, and a consistent approach applicable to the entire continental U.S. The speciation approach to emissions inventory development was chosen for this study because it was the best approach available for developing a comprehensive national modeling emissions inventory within budget and time constraints.

In 1997, EPA released its 1990 National Emission Trends (NET) inventory (U.S. Environmental Protection Agency 1997). This inventory is a revised version of the 1990 interim inventories for VOC and PM used in this study. National emissions totals in the NET inventory are lower than in the interim inventories by 33 percent for VOC emissions and 42 percent for PM. General reductions in VOC and PM emissions would suggest general reductions in the emissions of toxics estimated for this study. However, many of the large reductions in the VOC and PM emissions estimates are concentrated in source categories for which the interim inventory VOC and PM estimates were not used in this study. These categories include: chemical manufacturing and other manufacturing point sources (1990 TRI data were used instead for this study); waste treatment, storage and disposal facilities (alternate EPA data source (Coburn 1995) used instead for this study); and PM area source dust emissions for paved and unpaved roads, wind erosion construction, geogenic wind erosion, and agricultural crops tilling (emissions for these categories were not included in this study because of high uncertainties). In addition, the

NET inventory reflects increases of 36-46 percent, compared with the interim inventory used for this study, in PM emissions from point sources and nondust area sources. More detailed analysis of the differences between the interim inventory and the NET inventory, and their relationship to the emissions estimates used in this study, may be found in Chapter 3 of the modeling technical report (Rosenbaum et al. 1998).

Development of HAP emissions estimates from VOC and PM emissions estimates requires the application of speciation profiles. The primary source of many of these profiles is EPA's SPECIATE database (U.S. Environmental Protection Agency 1992). Previous studies have noted problems with specific profiles in the SPECIATE database (Harley et al. 1992; Korc and Chinkin 1993; Ligocki et al. 1992). As a result, profiles from SPECIATE were supplemented or revised using numerous other data sources in the technical literature. These profiles are detailed in Attachment 3 of the modeling technical report (Rosenbaum et al. 1998).

Some of the remaining uncertainties in the emissions inventory used in this study may be particularly important for the development of strategies for reducing risks from area sources. First, there are eight pollutants—1,1,2,2-tetrachloroethane, acrylamide, bis(2-ethylhexyl)phthalate; ethylene dibromide, hydrazine, methylene diphenyl diisocyanate, quinoline, and vinylidene chloride--with no area source emissions estimates in this study, but which do have area source emissions in the emissions inventory for 40 HAPs recently developed as part of the urban area source program. These area source emissions could result in exceedances of benchmark concentrations that have not been estimated in this

study. Second, this study's emissions inventory has relatively high contributions for area source chemical manufacturing and pharmaceutical manufacturing for several HAPs, including acrylonitrile, vinyl chloride, ethylene dichloride, chloroform, carbon tetrachloride, and ethyl acrylate. Both the VOC emissions estimates and the speciation profiles for these categories have large uncertainties--possibly resulting in the overestimation of benchmark concentration exceedances resulting from area source emissions for these HAPs.

The comparison of impacts from area sources, point sources, and mobile sources presented in this chapter also has uncertainties attributable to the definition of an "area source." As noted above, the area source definition used in constructing the emissions inventory for this analysis is not equivalent to the statutory definition of area source in CAA section 112. Some emissions treated as area source emissions in this analysis may actually be associated with section 112 major sources rather than section 112 area sources. An evaluation of the emissions inventory compiled for this study found that more than 70 percent of the estimated area source emissions are associated with categories which almost certainly are not CAA section 112 major sources, such as agricultural sources, dry cleaners, consumer and commercial products, and residential fuel combustion. However, significant portions of the area source emissions estimates for some other source categories, such as chemical manufacturing, petroleum refining, oil and gas production, and industrial surface coating, may be attributable to emitters which would be classified as major sources under section 112. Consequently, the relative contributions of

area sources to benchmark exceedances in this analysis may be greater than the contributions of emitters defined as area sources under section 112.

4.2 Modeled Concentrations

Comparison of modeled concentrations to the available monitoring data for air toxics indicates that the model estimates have an overall tendency to underestimate measured concentrations (Rosenbaum et al. 1998). A ratio of the modeled concentration to the monitored concentration was calculated for a total of 736 annual averaged monitored concentrations, obtained for 19 HAPs at 81 monitoring locations. The geometric mean of the set of model-monitor ratios is 0.53, and 73 percent of the ratios are less than one. As described in Section 2.1.4 above, only monitoring data sets with measurements below the minimum detection level (MDL, or “non-detect level”) totaling less than 10 percent of measurements taken in a year were used in this quantitative model performance evaluation.

It is possible that the findings of an overall tendency to underestimate are biased by the exclusion of measurement data sets dominated by observations below the non-detect level, since the data sets eliminated will tend to be those with lower concentrations. To test this hypothesis, additional model-monitor comparisons were conducted for the 13 HAPs measured in the Urban Air Toxics Monitoring Program with measurements dominated by individual observations below the non-detect level (this analysis is described in Attachment 5 of the modeling report (Rosenbaum et al. 1998)). For these non-detect data sets, the modeled concentration was compared to a range of possible concentrations, which was

calculated by assigning two values to each measurement below the non-detect level: a lower bound of zero, and an upper bound of the non-detect level itself.

For these more qualitative model-monitor comparisons, the modeled concentrations were lower than the lower bound of the possible monitored concentration range—an indication of model underestimation—in 57 percent of the 156 cases. The modeled concentrations were greater than the upper bound of the possible monitored concentration range—an indication of model overestimation—in 14 percent of the cases. The modeled concentrations were between the lower bound and the upper bound of the range in 29 percent of the cases, offering no clear information about the comparison between modeled and monitored concentrations. The high frequency of cases in which the modeled concentration is lower than the lower bound of the possible range of modeled concentrations is consistent with the conclusion that the general tendency is for the modeled concentrations to underestimate concentrations found by monitoring.

The tendency to underestimate outdoor concentrations of air toxics could result in underestimation of the frequency with which benchmark concentrations are exceeded. In addition, the modeled concentration outputs do not capture spatial or temporal peak concentrations that could be significant. The modeling approach used for this study estimates annual average concentrations at the census tract level. A census tract average concentration will not reflect areas within a census tract, such as locations close to a stationary source or a major roadway, which may have concentrations substantially greater than the average across the census tract. Also, a long-term average concentration will not reflect short-term elevated concentrations that may also have important health effects.

Consequently, a HAP concentration modeled in this study which is less than a benchmark concentration for the HAP does not mean that the benchmark is never exceeded within that census tract; consideration of alternate spatial and temporal scales could potentially identify additional benchmark exceedances.

In addition, the analysis in this chapter only presents the frequency with which benchmark concentrations are exceeded, and does not consider the magnitude of exceedance. A HAP which exceeds its benchmark concentration in a relatively small number of census tracts may exceed that benchmark by a large magnitude, and therefore may pose a greater potential risk than another HAP which exceeds its benchmark in more tracts but with a small magnitude of exceedance.

4.3 Benchmark Concentrations

For this study, a set of benchmark concentrations was compiled from a number of data sources, as described in Caldwell et al (Caldwell et al. 1998). Benchmark concentrations represent an estimated concentration at which a lifetime daily exposure is unlikely to result in adverse health effects, based on available hazard assessment data.

The benchmark concentration for cancer hazard is derived from the unit risk, an upper-bound estimate of the excess cancer risk over background incidence associated with a continuous lifetime exposure. Factors including use of sensitive animal strains, tumor sites of uncertain human relevance, and linear extrapolation to low doses can contribute to uncertainty in estimating the risk in human population (Cogliano 1997). Differences in the

pharmacokinetics of pollutants between exposure routes and species are expected, and can have influence on extrapolation of observed responses in animals and humans (U.S. Environmental Protection Agency 1994b).

This analysis emphasizes the inhalation route of exposure as benchmark concentrations were applied to modeled ambient air concentrations. However, health effects information is not always available for the inhalation route of exposure. For cancer benchmarks, extrapolations were needed to use available information from other routes of exposure. When extrapolating between two different routes of exposure (e.g., inhalation vs. oral), a number of factors are important for determining the association between a specific dose and the degree of toxic response engendered by a pollutant. These factors include differences by route of exposure in (1) tissue distribution, (2) rate of delivery leading to differing concentration profiles, (3) degree of metabolism, and (4) response caused by an agent at its site of action across species and among target tissue.

How such uncertainties affected the application of dose-response information for this type of analysis is not clear (U.S. Environmental Protection Agency 1994b).

However, in limited comparisons of differences between oral and inhalation dose routes, Pepelko concluded that the carcinogenic potencies are not substantially influenced by dose route (Pepelko 1990). However, the use of information extrapolated from oral to the inhalation route of exposure involves greater uncertainty than using that based on the inhalation route. This uncertainty is addressed by

assignment of cancer benchmark concentrations based on extrapolated data to Tier II rather than Tier I.

Benchmarks representing noncancer risks from long-term exposure make use of USEPA Reference Concentrations (RfCs) or similar values representing noncancer inhalation risks developed by other agencies. The RfC is by definition an estimate with an uncertainty spanning perhaps an order of magnitude. Although severity of effect is considered in the development of RfCs, there is no numerical adjustment for severity. Considerations of uncertainty are numerically represented in the derivation of RfCs to account for differences in human sensitivity, extrapolation from animals to humans, length of study, use of an observed rather than non-observed effect level, and completeness of the database. These uncertainties are addressed by use of conservative safety factors in derivation of the RfC; however, an RfC is not derived when it is determined that the uncertainties are too great (U.S. Environmental Protection Agency 1990).

In this study, only data on health effects via the inhalation route of exposure were used in establishing noncancer benchmark concentrations. No benchmarks for noncancer effects were developed through extrapolation from data for the oral route of exposure; oral studies are limited as indicators of non-cancer inhalation toxicity because of factors such as portal of entry effects and liver “first-pass effects,” as well as lack of consideration of dosimetric considerations (U.S. Environmental Protection Agency 1994b). For HAPs with no EPA inhalation RfCs, California EPA reference exposure

levels (RELS) and ATSDR minimal risk levels (MRLs) were used and assigned to Tier II.

Limitations in the availability of toxicity data for HAPs must be considered when assessing potential health impacts of these pollutants. Approximately 20 percent of the modeled HAPs with a weight of evidence indicating potential carcinogenicity do not have a cancer potency estimate and half do not have a benchmark concentration for noncancer health effects (Caldwell et al. 1998). Seventeen of the HAPs considered in this analysis have either an EPA weight-of-evidence determination as known, probable or possible human carcinogens or a recent National Toxicology Program study reporting clear evidence of animal carcinogenicity, but do not have carcinogenic potency estimates. For example, styrene is considered to be a possible (Group C) human carcinogen, but because it has no potency estimate, it was not possible to determine the frequency with which modeled styrene concentrations exceed a benchmark concentration. If styrene were assigned a default potency estimate that is consistent with other Group C carcinogens, modeled concentrations in a number of census tracts would exceed the benchmark concentration.

Even for some of the ubiquitous pollutants identified in this analysis, there is incomplete toxicity information. For example, benzene and 1,3-butadiene have both been associated with reproductive and developmental effects (U.S. Environmental Protection Agency 1994b), but currently have no benchmark concentration for such effects. In addition, 29 of the 148 HAPs included in this study have no Tier I or Tier II benchmark concentrations for any effects even though there are previous studies indicating some of these HAPs are

of potential health concern (U.S. Environmental Protection Agency 1994b). For example, n,n-dimethylaniline is ranked by EPA as being of high concern for noncancer effects, but quantitative hazard information is not available.

Another limitation in the toxicity information for the HAPs is in hazard evaluation for chemical groups. Outdoor concentrations were modeled for 14 HAP chemical groups. It is difficult to assess the toxicity of chemical groups, because they are comprised of a number of different species. For example, the HAP listed as “mercury compounds” is made up of several different constituents, including mercuric chloride, elemental mercury, mercuric nitrate, and mercury (aceto) phenyl, all with potentially different levels and types of toxicity. Also, the toxicity of the individual members of the polycyclic organic matter (POM) category varies significantly. This category is very broad and the toxicity of many of its members has not been characterized. However, many studies have shown the potential carcinogenic potency of polycyclic aromatic hydrocarbons—a subset of the POM category—to be large (U.S. Environmental Protection Agency 1993a). Assignment of an appropriate benchmark to this category depends on the extent to which particular POM constituents contribute to overall POM concentrations; differing assignments of hazard potential estimates for POM may profoundly affect estimates of the health risk posed by HAPs.

A further limitation of this analysis is that it only considers the potential health impact of individual pollutants. Additive or synergistic interactions among HAPs may pose a threat to public health beyond that identified in this chapter. HAP concentrations that are less

than benchmark concentrations may pose a risk to health when they are considered in combination with concentrations of other HAPs. Currently, too little is known about how pollutants interact to fully evaluate the potential health risks posed by exposure to multiple HAPs at concentrations below toxicity benchmarks.

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